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<b>(21) International Application Number:</b> PCT/US97/14213 <b>(22) International Filing Date:</b> 13 August 1997 (13.08.97)  <b>(30) Priority Data:</b> 60/024,799 13 August 1996 (13.08.96) US 08/891,112 10 July 1997 (10.07.97) US  <b>(71) Applicant:</b> E.I. DU PONT DE NEMOURS AND COMPANY [US/US]; 1007 Market Street, Wilmington, DE 19898 (US).  <b>(72) Inventors:</b> MERCHANT, Abid, Nazarali; 1408 Clive Circle, Wilmington, DE 19803 (US). MINOR, Barbara, Haviland; 233 Green Haven Drive, Elkton, MD 21921 (US). MOIYADI, Shoeb, Akberali; 47 Kings Grant Road, Hockessin, DE 19707 (US).  <b>(74) Agent:</b> KING, Karen, K.; E.I. du Pont de Nemours and Company, Legal/Patent Records Center, 1007 Market Street, Wilmington, DE 19898 (US).		<b>(81) Designated States:</b> AL, AM, AU, AZ, BA, BB, BG, BR, BY, CA, CN, CU, CZ, EE, GE, GH, HU, IL, IS, JP, KG, KP, KR, KZ, LC, LK, LR, LT, LV, MD, MG, MK, MN, MX, NO, NZ, PL, RO, RU, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UZ, VN, YU, ARIPO patent (GH, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report.</i>
<b>(54) Title:</b> ALKYL-SILOXANE COMPOSITIONS  <b>(57) Abstract</b>  Compositions containing alkylsiloxane and at least one compound selected from the group consisting of 1,1,1,2,3,4,4,5,5,5-decafluoropentane, nonafluoromethoxybutane, nonafluoroethoxybutane, dichloropentafluoropropane, 2,2-dichloro-1,1,1-trifluoroethane and isopropanol are described. These compositions are useful as cleaning agents, heat transfer media, and carrier fluids.		

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## ALKYLSILOXANE COMPOSITIONS

### CROSS-REFERENCE TO RELATED APPLICATION

This application claims the benefit of U.S. Provisional Application No.  
10 60/024,799 filed August 13, 1996.

### FIELD OF THE INVENTION

This invention relates to compositions containing alkylsiloxanes. These compositions include alkylsiloxane and at least one compound selected from the group  
15 consisting of 1,1,1,2,3,4,4,5,5,5-decafluoropentane, nonafluoromethoxybutane, nonafluoroethoxybutane, dichloropentafluoropropane, 2,2-dichloro-1,1,1-trifluoroethane and isopropanol. These compositions are useful as cleaning agents, heat transfer media, and carrier fluids.

### BACKGROUND OF THE INVENTION

20 Fluorinated hydrocarbons have many uses such as cleaning agents, drying agents or carrier fluids. Such compounds include 1,1,2-trichloro-1,2,2-trifluoroethane (CFC-113). In recent years it has been pointed out that certain kinds of fluorinated hydrocarbons released into the atmosphere may adversely affect the stratospheric ozone  
25 layer. Although this proposition has not yet been completely established, there is a movement toward the control of the use and production of certain chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs) under an international agreement. There is a parallel concern about the contribution perfluorinated compounds make to global warming, therefore, it is desirable to find compounds that are partially fluorinated to  
30 reduce potential for global warming.

Accordingly there is a demand for the development of new compounds that have lower ozone depletion potential and lower global warming contribution than existing compounds while still achieving acceptable cleaning performance.

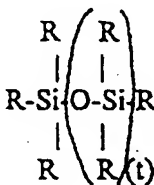
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5 It is desirable to find partially fluorinated compounds for use as a cleaning agent to clean for example, silicone off medical instruments and tubing. Partially fluorinated compounds may also be useful as a carrier fluid, for example, to place a thin layer of silicone or another compound on a surface. To clean a surface containing silicone or to place silicone on a surface, it is desirable to add compounds to the partially  
 10 fluorinated solvent to enhance solubility of silicone. It is particularly desirable that these compounds be non-flammable. These enhanced solvents may also be useful as heat transfer fluids, particularly in secondary loop systems. Enhanced solvents can also act as carriers for other compounds such as adhesion promoter accelerators, initiators and catalysts.

15 Accordingly, it has been found that adding specific agents to partially fluorinated compounds can enhance solubility of residues, for example, silicone, enhance cleaning performance, act as carrier fluids and heat transfer fluids.

### SUMMARY OF THE INVENTION

20 The present invention relates to the following compositions: a first component, alkylsiloxane of the formula



25 where R is an alkyl group having from 1 to 10 carbon atoms and t is an integer from 1 to 5, and a second component, wherein the second component is selected from the group consisting of 1,1,1,2,3,4,4,5,5,5-decafluoropentane, nonafluoromethoxybutane, nonafluoroethoxybutane and dichloropentafluoropropane and optionally, a third  
 30 component, wherein the third component is selected from the group consisting of isopropanol and 2,2-dichloro-1,1,1-trifluoroethane.

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These compositions may be useful as cleaning agents, heat transfer media, silicone removal agents, and as carrier fluids for silicone, adhesion promoters or other compounds.

### DETAILED DESCRIPTION

10 The present invention relates to the discovery of mixtures of alkylsiloxane and at least one of 1,1,1,2,3,4,4,5,5-decafluoropentane, nonafluoromethoxybutane, nonafluoroethoxybutane, dichloropentafluoropropane, isopropanol and 2,2-dichloro-1,1,1-trifluoroethane.

15 1-99% of each of the above components can be used as cleaning agents, heat transfer media, silicone removal agents, and as carrier fluids for compounds such as silicone, adhesion promoters or other compounds.

Nonafluoromethoxybutane ( $C_4F_9OCH_3$ ) isomers of the present invention include 1,1,1,2,2,3,3,4,4,-nonafluoro-4-methoxy-butane ( $CH_3OCF_2CF_2CF_2CF_3$ ), 1,1,1,2,3,3,-hexafluoro-2-(trifluoromethyl)-3-methoxy-propane ( $CH_3OCF_2CF(CF_3)_2$ ), 20 1,1,1,3,3,3-hexafluoro-2-methoxy-2-(trifluoromethyl)-propane ( $CH_3OC(CF_3)_3$ ), and 1,1,1,2,3,3,4,4,4-nonafluoro-2-methoxy-butane ( $CH_3OCF(CF_3)CF_2CF_3$ ), approximate isomer boiling point =  $60^\circ C$ ;

Nonafluoroethoxybutane ( $C_4F_9OC_2H_5$ ) isomers of the present invention include 1,1,1,2,2,3,3,4,4-nonafluoro-4-ethoxybutane ( $CH_3CH_2OCF_2CF_2CF_2CF_3$ ), 25 1,1,1,2,3,3-hexafluoro-2-(trifluoromethyl)-3-ethoxypropane ( $CH_3CH_2OCF_2CF(CF_3)_2$ ), 1,1,1,3,3,3-hexafluoro-2-ethoxy-2-(trifluoromethyl)-propane ( $CH_3CH_2OC(CF_3)_3$ ), and 1,1,1,2,3,3,4,4,4-nonafluoro-2-ethoxybutane ( $CH_3CH_2OCF(CF_3)CF_2CF_3$ ) with approximate isomer boiling points of  $73^\circ C$ ;

Dichloropentafluoropropane isomers include 1,1-dichloro-2,2,3,3,3-pentafluoropropane ( $CHCl_2CF_2CF_3$ , HCFC-225ca), boiling point =  $50^\circ C$ , and 30 1,3-dichloro-1,1,2,2,3-pentafluoropropane ( $CHClCF_2CClF_2$ , HCFC-225cb), boiling point =  $56^\circ C$ . 2,2-dichloro-1,1,1-trifluoroethane (HCFC-123 or  $CHCl_2CF_3$ ) has a boiling point of  $27^\circ C$ . Isopropanol ( $(CH_3)_2CHOH$ ) has a boiling point of  $82.2^\circ C$ .

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5 Effective amounts of these compositions can contain from 10 to 60 weight percent alkylsiloxane and 40 to 90 weight percent of at least one of HFC-43-10mee,  $C_4F_9OCH_3$ ,  $C_4F_9OC_2H_5$ , HCFC-225ca, or HCFC-225cb. Effective amounts can also contain 1-30 weight percent alkylsiloxane, 9-50 weight percent HCFC-123, and 25-90 weight percent of at least one of HFC-43-10mee,  $C_4F_9OCH_3$ ,  $C_4F_9OC_2H_5$ , HCFC-225ca, 10 or HCFC-225cb. Compositions may also contain effective amounts of 10-59 weight percent alkylsiloxane, 1-20 weight percent isopropanol, and 40-89 weight percent of at least one of HFC-43-10mee,  $C_4F_9OCH_3$ ,  $C_4F_9OC_2H_5$ , HCFC-225ca, or HCFC-225cb.

Specific examples illustrating the invention are given below. Unless otherwise stated therein, all percentages are by weight. It is to be understood that these 15 examples are merely illustrative and in no way are to be interpreted as limiting the scope of the invention.

#### EXAMPLE 1

##### Flammability Test

20 A Penski-Martin Closed Cup flash point tester was filled with mixtures shown in Table 1. OS-10 is hexamethylsiloxane and OS-20 is octamethyltrisiloxane. Flash points were determined in a temperature range from about -20°C to 38°C. No flash points were observed at any temperature for any mixture tested. Flammability was also tested by attempting to ignite mixtures in a pan with a spark and a flame. No ignitions 25 resulted in mixtures tested.

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TABLE 1

Flammability Data

	Mixture Wt%	Flash Point	Ignition in Pan
	HFC-43-10mee/OS-10		
	80/20		None
10	70/30		None
	60/40		None
	50/50		None
	HFC-43-10mee/HCFC-123/OS-10		
	50/30/20	None	None
15	60/20/20		None
	55/25/20		None
	60/25/15		None
	45/35/20	None	None
	45/30/25	None	None
20	HFC-43-10mee/OS-20/Isopropanol		
	50/40/10		None
	60/30/10		None
	70/20/10		None

25

EXAMPLE 2

A suitable container was filled with mixtures shown in Table 2 and mixed thoroughly at room temperature. Trans-1,2-DCE is trans-1,2-dichloroethylene (CCl=CCl). Dow Corning 360, Dow Corning 550, or Dow Corning 1107 medical grade silicone oil was then slowly added to each mixture until the blend would no longer dissolve the oil.

30

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TABLE 2

	<u>Mixture</u>	<u>Weight Percent</u>	<u>Wt. % Silicone Dissolved</u>		
			DC-360	DC-550	DC-1107
	CFC-113	100	16.0		
	Hexane	100	16.0		
10	43-10mee/ cyclopentane/ trans-1,2-dichloroethylene	65/15/20	0.5		
	43-10mee/ cyclohexane/ trans-1,2-dichloroethylene	85/5/10	0.5		
15	43-10mee/OS-10	60/40	10.02		
		50/50	23.0		
	43-10mee/123/OS-10	50/30/20	5.12	14.0	25.0
		60/25/15	<1.0	1.68	9.0
20		60/20/10	<1.0	3.01	9.0
		47.5/35/17.5	5.0	12.0	24.0
		45/30/25	18.2		
		50/35/15	5.5		
		45/35/20	12.85		
25	43-10mee/OS-20/ isopropanol	50/40/10	3.5		

Results show that the addition of hexamethyldisiloxane to HFC-43-10mee significantly improves silicone solubility. Alkysyloxane-containing blends can also act as silicone carrier fluids due to their ability to dissolve silicone. These blends also have the advantage of being non-flammable and non-toxic.

EXAMPLE 3

A suitable container was filled with the mixtures shown in Table 3. Pre-weighed tubing made of polyurethane was immersed in each mixture at a specified temperature for three minutes. Parts were then dried, weighed and observed for swelling.



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TABLE 3

Weight Gain and Enlargement

	Pre-Test	Post-Test		% Wt	Change in	
	<u>Solvent</u>	<u>Wt (kg)</u>	<u>Wt (kg)</u>	<u>BP(°C)</u>	<u>Increase</u>	<u>Dimension</u>
10	CFC-113	0.1917	0.213	47	11.1	Enlarged
	HFC-43-10mee/ cyclopentane/ trans-1,2-DCE (65/15/20 wt%)	0.174	0.2209	37	27.0	Enlarged and softened
15	Hexane	0.1669	0.2112	68	26.5	Enlarged
	HFC-43-10mee/ 123/OS-10 (85/15 wt%)	0.1481	0.1481	55.5	0.0	No effect

Results show the blends containing alkylsiloxanes demonstrated significantly reduced weight gain and swelling versus the other compositions tested.

EXAMPLE 4

Polyurethane, nylon and ABS tubing were coated with silicone and then cleaned with a solution containing 85 weight percent HFC-43-10mee and 15 weight percent hexamethyldisiloxane. Results are shown in Table 4 below.

TABLE 4

Cleanability

<u>Tubing Type</u>	<u>Degree of Cleaning</u>
Polyurethane	Visually Clean
ABS	Visually Clean
Nylon	Visually Clean

Tubing also felt clean with no evidence of silicone remaining on the tubes.

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5

## EXAMPLE 5

Lengths of Polysilicone tubing (Boston Scientific) were cut with tube weight and diameter recorded. A suitable container was filled with solvent compositions as shown in Table 5. Each composition was heated to the vapor temperatures shown and tubing was immersed in the solvent for several minutes. Tubing was removed and allowed to dry for 30 seconds. Tubing was then reweighed and the diameter remeasured. The change in weight and diameter were calculated.

TABLE 5

		Change in Weight (g)	Change in Diameter(in)	Vapor T(°C)	
15	Wt%				
	3 minute immersion:				
	HCFC-225	100	+0.123	+0.016	53.5
	CFC-113	100	+0.1738	+0.024	47.5
	HFC-43-10mee/OS-10	50/50	+0.0778	+0.023	57.0
20	HFC-43-10mee/OS-20	50/50	+0.0942	+0.031	57.0
	HCFC-225	100	+0.1182	+0.016	21.0
	CFC-113	100	+0.0874	+0.020	"
	HFC-43-10mee/OS-10	50/50	+0.0713	+0.018	"
	HFC-43-10mee/OS-20	50/50	+0.0611	+0.023	"
25	15 minute immersion:				
	CFC-113	100	+0.3710	+0.038	21.0
	HFC-43-10/123/OS-10	50/30/20	+0.1595	+0.026	"
	HFC-43-10/OS-20/IPA	50/40/10	+0.1235	+0.023	"
	HFC-43-10/OS-20/IPA	60/20/10	+0.1031	+0.021	"
30	HFC-43-10/OS-20/IPA	70/20/10	+0.0871	+0.019	"

For connecting polysilicone tubing using a solvent, it is desirable to have a significant increase in tube diameter without a large increase in tube weight. The samples containing alkylsiloxanes generally showed adequate change in tube diameter and the smallest increase in tube weight.

ADDITIONAL COMPOUNDS

Other components, such as aliphatic hydrocarbons having a boiling point of 0-100°C, hydrofluorocarbonalkanes having a boiling point of 0-100°C, hydrofluoropropanes having a boiling point of between 0-100°C, hydrocarbon esters having a boiling point between 0-100°C, hydrochlorofluorocarbons having a boiling point between 0-100°C, hydrofluorocarbons having a boiling point of 0-100°C, hydrochlorocarbons having a boiling point between 0-100°C, chlorocarbons and perfluorinated compounds, can be added to the azeotropic or azeotrope-like compositions described above without substantially changing the properties thereof, including the constant boiling behavior, of the compositions. Examples of such components, which typically do not exceed about 10 weight percent of the total composition, include the following:

	<u>COMPOUND</u>	<u>FORMULA</u>	<u>boiling point, °C</u>
20	HCFC-123	$\text{CHCl}_2\text{CF}_3$	27
	HCFC-141b	$\text{CFCl}_2\text{CH}_3$	32
	HCFC-225aa	$\text{CHF}_2\text{CCl}_2\text{CF}_3$	53
	HCFC-225da	$\text{CClF}_2\text{CHClCF}_3$	50
	HFC-HFC-43-10mf	$\text{CF}_3\text{CH}_2\text{CF}_2\text{CF}_2\text{CF}_3$	52
25	HFC-HFC-43-10mcf	$\text{CF}_3\text{CF}_2\text{CH}_2\text{CF}_2\text{CF}_3$	52
	FC-C-51-12	$\text{cyclo-C}_4\text{F}_6(\text{CF}_3)_2$	45
		$\text{CH}_3\text{OCF}_2\text{CHF}_2\text{CF}_3$	52
	HFC-C-354	$\text{cyclo-CF}_2\text{CF}_2\text{CH}_2\text{CH}_2$	50
		$\text{C}_4\text{F}_9\text{CH}=\text{CH}_2$	58
30	MEK	$\text{CH}_3\text{C}(\text{O})\text{C}_2\text{H}_5$	80
	THF	$\text{cyclo-OC}_4\text{H}_8$	66
	methyl formate	$\text{HC}(\text{O})\text{OCH}_3$	32
	ethyl formate	$\text{HC}(\text{O})\text{OC}_2\text{H}_5$	54
	methyl acetate	$\text{CH}_3\text{C}(\text{O})\text{OCH}_3$	56
35	ethyl acetate	$\text{CH}_3\text{C}(\text{O})\text{OC}_2\text{H}_5$	77
	1,2-dichloroethane		84
	acetonitrile		82
	methylene chloride		40
	heptane	$\text{CH}_3(\text{CH}_2)_5\text{CH}_3$	98

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Additives such as lubricants, corrosion inhibitors, stabilizers, surfactants, dyes and other appropriate materials may be added to the novel compositions of the invention for a variety of purposes provided they do not have an adverse influence on the composition, for their intended applications. Examples of stabilizers include nitromethane and nitroethane.

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CLAIMSWhat is claimed is:

1. A composition comprising effective amounts of alkylsiloxane and at  
10 least one compound selected from the group consisting of 1,1,1,2,3,4,4,5,5,5-  
decafluoropentane, nonafluoromethoxybutane, nonafluoroethoxybutane,  
dichloropentafluoropropane, 2,2-dichloro-1,1,1-trifluoroethane and isopropanol.
2. The composition of claim 1 comprising 10-60 weight percent  
15 alkylsiloxane and 40-90 weight percent 1,1,1,2,3,4,4,5,5,5-decafluoropentane; 10-60  
weight percent alkylsiloxane and 40-90 weight percent nonafluoromethoxybutane; 10-60  
weight percent alkylsiloxane and 40-90 weight percent nonafluoroethoxybutane; 10-60  
weight percent alkylsiloxane and 40-90 weight percent 1,1-dichloro-2,2,3,3,3-  
pentafluoropropane; 10-60 weight percent alkylsiloxane and 40-90 weight percent 1,3-  
20 dichloro-1,1,2,2,3-pentafluoropropane; 10-60 weight percent alkylsiloxane, 1-89 weight  
percent 1,1-dichloro-2,2,3,3,3-pentafluoropropane and 1-89 weight percent 1,3-dichloro-  
1,1,2,2,3-pentafluoropropane.
3. The composition of Claim 1 comprising 1-30 weight percent  
25 alkylsiloxane, 9-50 weight percent 2,2-dichloro-1,1,1-trifluoroethane, and 25-90 weight  
percent of at least one compound selected from the group consisting of  
1,1,1,2,3,4,4,5,5,5-decafluoropentane, nonafluoromethoxybutane,  
nonafluoroethoxybutane, 1,1-dichloro-2,2,3,3,3-pentafluoropropane and 1,3-dichloro-  
1,1,2,2,3-pentafluoropropane.
- 30 4. The composition of Claim 1 comprising 10-59 weight percent  
alkylsiloxane, 1-20 weight percent isopropanol, and 40-89 weight percent of at least one  
compound selected from the group consisting of 1,1,1,2,3,4,4,5,5,5-decafluoropentane,

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5 nonafluoromethoxybutane, nonafluorooctoxybutane, 1,1-dichloro-2,2,3,3,3-pentafluoropropane and 1,3-dichloro-1,1,2,2,3-pentafluoropropane.

5. A composition comprising 1-30 weight percent hexamethyldisiloxane, 25-90 weight percent 1,1,1,2,3,4,4,5,5,5-decafluoropentane and 9-50 weight percent 2,2-  
10 dichloro-1,1,1-trifluoroethane; and 10-59 weight percent octamethyltrisiloxane, 40-89 weight percent 1,1,1,2,3,4,4,5,5,5-decafluoropentane and 1-20 weight percent isopropanol.

15 6. A process for cleaning a solid surface which comprises cleaning said surface with a composition of claim 1 or 5.

7. A process for carrying an active ingredient in a solvent in which the solvent comprises a composition of claim 1 or 5.

20 8. A process for transferring heat from a heat source to a heat sink using a composition of claim 1 or 5.

9. A fluid for carrying silicone comprising a composition of claim 1 or 5.

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## INTERNATIONAL SEARCH REPORT

International Application No.

PL/US 97/14213

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C11D7/50 C11D7/26 C11D7/28

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C11D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 699 746 A (KABUSHIKI KAISHA TOSHIBA) 6 March 1996 see page 24; example 29; table 10 see page 25; example 30; table 11 see page 11, line 50 - page 12, line 2 ---	1,2,6,7
X	EP 0 576 687 A (OLYMPUS OPTICAL CO. LTD.) 5 January 1994 see page 10 - page 11; examples 7,8; table 4 ---	1,6,7
A	EP 0 710 715 A (AG TECHNOLOGY CO. LTD.) 8 May 1996 see page 1, line 1 - line 38 see page 3, line 50 - line 51 see page 4, line 26 - line 34 --- -/-	1

☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

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## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>DATABASE WPI  Week 9614  Derwent Publications Ltd., London, GB;  AN 96-134156[37]  XP002046160  "Aq. solution containing  polyorganosiloxane and fluoro-compounds"  &amp; JP 08 024 806 (OLYMPUS OPTICAL CO. LTD.)  , 30 January 1996  see abstract  &amp; JP 08 024 806 A</p> <p>---</p>	1,6
A	<p>DATABASE WPI  Week 9423  Derwent Publications Ltd., London, GB;  AN 94-186550[23]  XP002046161  "Cleaning composition useful as  alternative for freon(s) - comprises a  per:fluorocarbon and a low m.wt. siloxan"  &amp; JP 06 122 898 (OLYMPUS OPTICAL CO. LTD.)  , 6 May 1994  see abstract  &amp; JP 06 122 898 A</p> <p>---</p>	1,6
X	<p>DATABASE WPI  Week 9403  Derwent Publications Ltd., London, GB;  AN 94-023141[03]  XP002046162  "finishing liquid for manual wiping"  &amp; JP 05 331 494 (OLYMPUS OPTICAL CO. LTD.)  , 14 December 1993  see abstract  &amp; JP 05 331 494 A  see page 2, formula 1</p> <p>---</p>	1,6,7
A	<p>EP 0 717 129 A (KABUSHIKI KAISHA TOSHIBA)  19 June 1996  see claims; examples</p> <p>---</p>	1,6
A	<p>WO 94 23008 A (MINNESOTA MINING &amp;  MANUFACTURING COMPANY) 13 October 1994  see page 3, line 14 - page 4, line 12;  claim 9; examples 9,10</p> <p>---</p>	1,6
A	<p>US 2 993 866 A (VAUGHN; WELLMAN) 25 July  1961  see examples</p> <p>-----</p>	1,6,7



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